Measuring Mercury Trends in Freshwater Fish, 2012 Sampling Results

DEPARTMENT OF ECOLOGY
State of Washington

Callie Mathieu, Michael Friese, and Andy Bookter, Environmental Assessment Program

2012 Highlighted Findings

- Mercury levels in bass increased in Deer (+18%), Fazon (+13%), and St. Clair (+28%) Lakes since 2007.
- Mercury levels increased 78% in Lake Samish bass between 2001-2012.
- Mercury levels decreased 24% in Lower Goose Lake bass since 2007.
- Mercury levels did not change in Lake Ozette bass between 2007-2012.

Why Does it Matter?

Mercury is highly toxic and bioaccumulative. Levels increase up aquatic food chains, resulting in concentrations among top predator fish that can be harmful to fish-eating wildlife and humans. As a neurotoxin, mercury can harm the brain and nervous system.

Overview

The Washington State Department of Ecology (Ecology) began a monitoring program in 2005 with the primary goal of assessing temporal trends in mercury levels of freshwater fish throughout the state. Each year, Ecology collects 10 individual largemouth or smallmouth bass from 6 lakes for analysis of total mercury. Ecology returns to each set of lakes every 5 years to assess trends and determine if mercury levels are changing. Additional species are also collected and analyzed as composites for a secondary goal of supporting fish consumption advisories.

In 2012, Ecology again sampled fish from Deer, Fazon, Lower (L.) Goose, Ozette, Samish, and St. Clair Lakes (Figure 1).

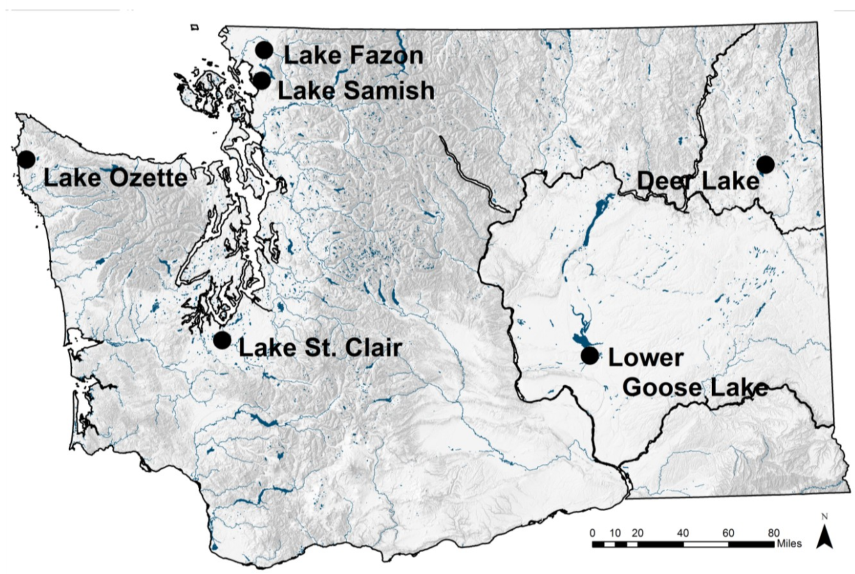

Figure 1. Locations of 2012 Sampling Sites.

This monitoring program supports Ecology and the Washington State Department of Health (DOH) in their efforts to reduce mercury in Washington's environment. Together, the departments developed a chemical action plan (CAP) for mercury in 2003 (Peele et al., 2003). The CAP addressed the threat of mercury in Washington State and made recommendations for state actions to reduce mercury.

For More Information

PBT Monitoring Program website: **www.ecy.wa.gov/programs/eap/toxics/pbt.html**

Chemical Action Plan website: **www.ecy.wa.gov/programs/swfa/pbt/caps.html**

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Methods

In the fall of 2012, Ecology collected 77 individual bass and 115 fish of additional species to form 25 composite samples. Bass were analyzed individually to achieve statistical power in detecting trends. Composites of additional species help inform DOH's fish consumption advisories. Fish were collected and processed following Ecology standard operating procedures (Sandvik, 2010a and 2010b). A summary of sampling conducted in 2012 is provided in Table 1.

Table 1. Summary of Sampling Conducted in 2012.

Collection Goal	Deer	Fazon	L Goose	Ozette	Samish	St. Clair
10 individual bass	+[1]	+	+[1]	+[2]	+	+
Species collected for composites	BBH, RBT	BBH, BG	LWF	NPM, PEA, YP	NPM, SMB, YP	RKB, BG, YP
2 water samples	+	+	+	+	+	+
Multi-parameter profile	+	+	+	+	+	+

"+": goal achieved. [1]10 smallmouth bass and 10 largemouth bass collected. [2]7 largemouth bass collected in 2012 and 3 collected in 2011. BBH: brown bullhead; RBT: rainbow trout; BG: bluegill; LWF: lake whitefish; NPM: northern pikeminnow; PEA: peamouth; YP: yellow perch; SMB: smallmouth bass; RKB: rock bass.

Fish collection efforts at Lake Ozette yielded only 7 bass in 2012. Three bass (ages < 4 year old) from Ozette were collected and analyzed for mercury in 2011; these samples were included in this report as part of the "2012" dataset.

Edible muscle tissue from fish were analyzed individually or as 3 to 5 fish composite samples for total mercury using cold vapor atomic absorption (EPA Method 245.6). Data generally met measurement quality objectives outlined in the Quality Assurance Project Plan (QAPP) (Seiders, 2006) and were deemed usable without qualification. Detailed quality assurance data are available upon request.

To better understand patterns, dynamics, and changes in mercury accumulation in fish, Ecology also collects water chemistry and in-situ measurements from each site. Figure 2 displays all parameters collected or measured at each site in 2012.

More detailed information on the study design of this project can be found in the QAPP (Seiders, 2006) and QAPP Addendum (Meredith and Furl, 2010).

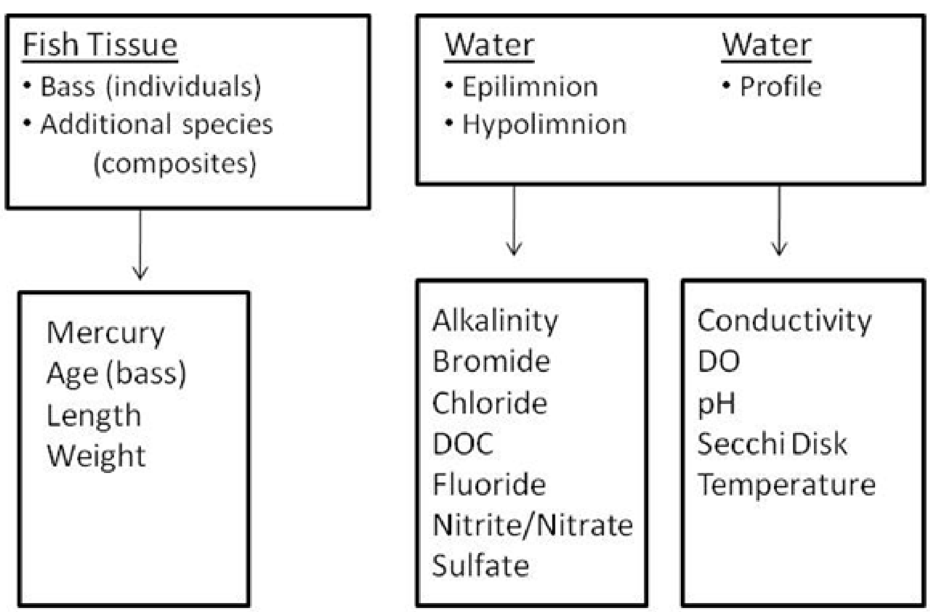


Figure 2. Analytes and Measurements Recorded at each Waterbody.
DOC: dissolved organic carbon; DO: dissolved oxygen.

Fish Tissue Criteria

The 1992 National Toxic Rule (NTR) and the 2001 EPA Recommended Criterion provide numerical thresholds useful in assessing mercury levels of waterbodies. Ecology adopted the NTR Rule as the state's regulatory criterion to determine whether a waterbody meets water quality standards in Washington State. For freshwater, the NTR fish tissue criterion is 770 ppb. The EPA Recommended Criterion sets the threshold at 300 ppb. This report compares fish tissue data to both criteria.

Ecology's role is to determine whether water quality standards are met and to begin the process to correct problems where standards are not met. DOH and local health departments are responsible for developing fish consumption advisories in Washington. DOH uses the mercury data from this monitoring program and other programs at Ecology to establish and update fish consumption advisories. For more information on DOH's fish consumption advisories, visit www.doh.wa.gov/CommunityandEnvironment/Food/Fish.aspx.

Waterbody Descriptions

All 2012 lakes were previously sampled in 2007 (Furl and Meredith, 2008). Lake selection in 2007 considered the following criteria: popularity among anglers, availability of target fish species, and inclusion in a 2001-2002 statewide bass survey (Fischnaller et al., 2003). The lakes cover a range of watershed characteristics, from wet and forested (Lake Ozette) to dry and agricultural or barren (L. Goose). Precipitation, forest cover, and agricultural land use have been identified as explanatory factors in fish tissue mercury levels in Washington State (Mathieu et al., 2013). Physical and morphological data on the sampling locations are provided in Furl and Meredith (2008).

Water Sample Results

Profiles of temperature, dissolved oxygen (DO), and pH indicated thermal stratification in all of the lakes, as well as a drop in DO near the bottom at all except for Lake Ozette (where bottom measurements were not reached) at the time of sampling. pH levels were highest in L. Goose Lake and lowest in Ozette and Samish Lakes.

Epilimnion (upper) and hypolimnion (lower) water samples showed high levels of alkalinity and sulfate in L. Goose Lake and greater concentrations of chloride and DOC in Lake Fazon (Figure 3). Fluoride and nitrite/nitrate results were generally low or not detected.

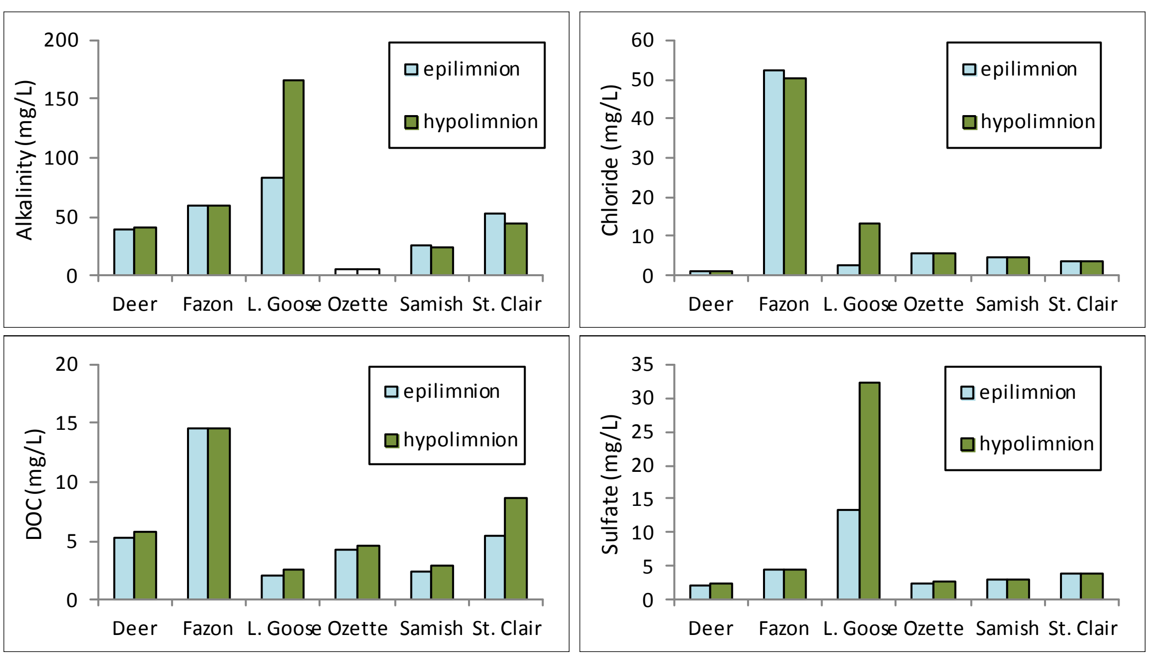


Figure 3. Alkalinity, Chloride, DOC, and Sulfate Concentrations in Water Samples. *Unfilled bars (Lake Ozette alkalinity) indicate parameter was not detected at that level.*

2012 Results for Mercury in Fish

Individual Fish Samples (Bass)

Results of 2012 mercury and water chemistry data are available for download at www.ecy.wa.gov/eim by searching Study ID: HgFish12. Mercury concentrations of individual smallmouth and largemouth bass are shown in Figure 4 and summary statistics are presented in Table 3. Mercury concentrations in individual bass ranged from 81 - 830 ppb. Levels were generally highest in Lake St. Clair largemouth bass and lowest in L. Goose Lake smallmouth bass.

Of the individual bass analyzed, 55% contained mercury levels higher than the 300 ppb EPA recommended criterion. Two bass samples—from Lakes Ozette and St .Clair— exceeded the National Toxics Rule (NTR) regulatory criterion of 770 ppb. The Lake Ozette sample was collected from a 4-year-old male measuring 400 mm, and the St. Clair sample from an 11-year-old female (length = 493 mm). Levels in 2 other bass from Lake St. Clair were close to the NTR, at 765 and 767 ppb.

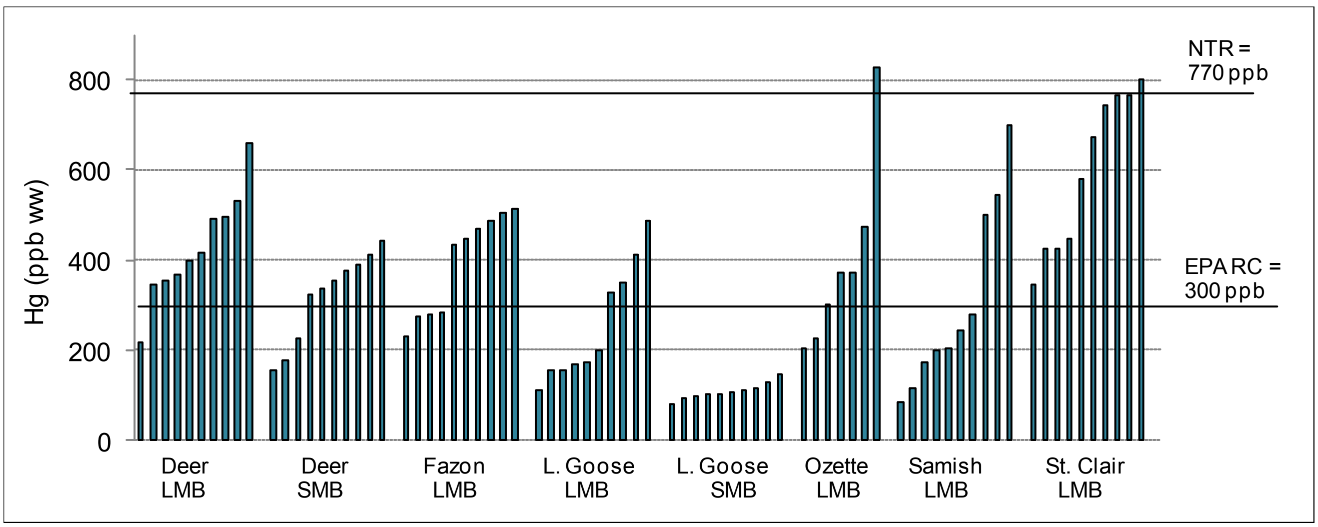


Figure 4. Mercury Concentrations in Individual Fish Collected in 2012. *NTR: National Toxics Rule; EPA RC: EPA Recommended Criterion; LMB: largemouth bass; SMB: smallmouth bass.*

Table 3. Summary Statistics of Bass Lengths, Weights, Ages, and Mercury Concentrations.

Waterbody	Species	Length (TL mm)		Weight (g)		Age (yr)		Hg (ppb)	
		Range	Mean	Range	Mean	Range	Mean	Range	Mean
Deer	LMB	323 - 480	380	598 - 1660	982	3 - 9	5.8	215 - 658	427
Deer	SMB	263 - 337	292	214 - 499	302	3 - 6	5.2	157 - 442	319
Fazon	LMB	263 - 442	385	258 - 1470	812	3 - 10	5.9	228 - 515	440
Lower Goose	LMB	350 - 492	406	737 - 2107	1196	3 - 9	4.8	112 - 448	254
Lower Goose	SMB	233 - 395	294	188 - 1072	444	1 - 3	1.7	81 - 147	108
Ozette	LMB	263 - 400	298	266 - 1128	459	2 - 4	2.7	203 - 830	397
Samish	LMB	201 - 410	279	103 - 1025	350	1 - 7	2.5	85 - 701	304
St. Clair	LMB	263 - 493	397	242 - 2062	1094	3 - 11	7.4	347 - 802	598
All Sites	---	201 - 493	340	103 - 2107	714	1 - 11	4.6	81 - 830	348

2012 Results for Mercury in Fish

Composite Fish Samples

Mercury levels in composite fish samples ranged from 94 - 2,580 ppb, with a median of 245 ppb (Figure 5). Composite samples from Lake Ozette and Lake Samish contained the highest levels of mercury. Concentrations were relatively low in composites from Deer, Fazon, L. Goose, and St. Clair Lakes.

All Ozette samples were above the EPA recommended criterion and 3 samples exceeded the NTR regulatory criterion. One northern pikeminnow sample contained the highest level of mercury yet recorded in Ecology's Environmental Information Management (EIM) database (2,580 ppb), which includes all fish caught for the Mercury Trends program since 2005, as well as all fish analyzed by Ecology. The fish included in the composite were fairly large, with an average length of 515 mm.

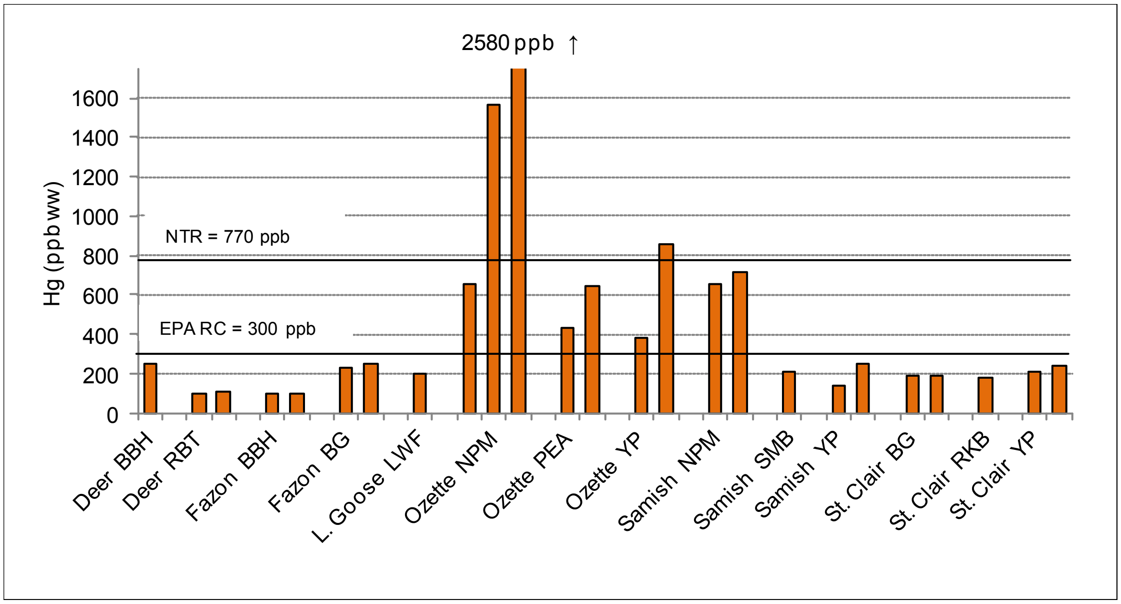


Figure 5. Mercury Concentrations of Composite Fish Samples Collected in 2012.
NTR: National Toxics Rule; EPA RC: EPA Recommended Criterion; BBH: brown bullhead; RBT: rainbow trout; BG: bluegill; LWF: lake whitefish; NPM: northern pikeminnow; PEA: peamouth; YP: yellow perch; SMB: smallmouth bass; RKB: rock bass.

Two Lake Samish northern pikeminnow composites also exceeded the EPA recommended criterion; while yellow perch and bluegill from the lake did not. No Samish composites exceeded the NTR regulatory criterion.

Fish Tissue Mercury Relationships

Mercury concentrations in bass significantly increased with fish size and age at all lakes sampled in 2012. In Deer Lake, mercury relationships with weight and age were significant, but not length. One fish from Deer Lake was outside of the normal length-weight relationship and had a Fulton's Fish Condition index score of 3.43, skewing the mercury relationship.

Starting in 2010, water samples have been analyzed for sulfate and other ions, as well as alkalinity and DOC. For this 2010-2012 data set, no significant relationships were found between mercury levels in bass and lake water chemistry. Future sampling may reveal significant correlations by increasing sample size.

While not statistically significant correlations, bass collected in 2012 had the highest mercury concentrations at the lakes with the lowest alkalinity and ion levels (Ozette and Samish), while the bass with lowest mercury levels came from the high alkalinity lake (L. Goose). The first 5 years of sampling for this program revealed that mercury levels in fish were higher in lakes with lower alkalinity and higher epilimnion temperatures (Mathieu et al., 2013).

Temporal Trends

Temporal trends in bass mercury levels were assessed in two ways: (1) using analysis of covariance (ANCOVA) to determine if statistically significant differences existed between collection years, and (2) calculating percent change of mercury levels in standard-size bass to relate how much levels have increased or decreased.

Individual bass were analyzed for mercury in 2007 and 2012 at all 6 lakes. Additional data from a 2001 survey were available for Lake Samish (Fischnaller et al., 2003). The 2001 data were used with a correction factor applied to adjust for differences in analytical methods (Furl, 2007a). Fish over 400 mm in length were excluded from the 2001 data set, as the correction factor is robust until that point. The Fazon Lake data set contained unequal regression slopes and was constrained to include only fish within the same size range. An outlying datapoint was excluded from the Deer Lake 2012 data set (studentized residual = 4.468). After exclusion, the mercury-length relationship was significant and linear. This sample was discussed in the previous section on relationships.

ANCOVA

Analysis of covariance (ANCOVA) with Bonferroni post-hoc tests were conducted to assess significant differences in $\log_{10}$ mercury levels in bass between collection years. Log_{10} fish length was used as the covariate with most of the lake data sets to control for the effect of fish size on mercury accumulation. Log_{10} fish age was used as the covariate for Lake St. Clair because the two collection years showed differing mercury-length relationship slopes, but fish age did not. No covariate was used for Lake Fazon, as no linear relationships were significant in the constrained data set. All assumptions for ANCOVA were verified prior to analysis.

Length-adjusted mercury means (least squares mean of $\log_{10}$ data) were significantly higher in Deer, Fazon, Samish, and St. Clair bass collected in 2012 than in bass collected in 2007—or in 2001 for Lake Samish only. Mercury means were significantly lower in the L. Goose Lake 2012 bass. No difference was found in mercury means of the Lake Ozette bass. Figure 6 shows the least square means resulting from the ANCOVA and Bonferroni post-hoc tests, along with ANCOVA F-ratios and significance values.

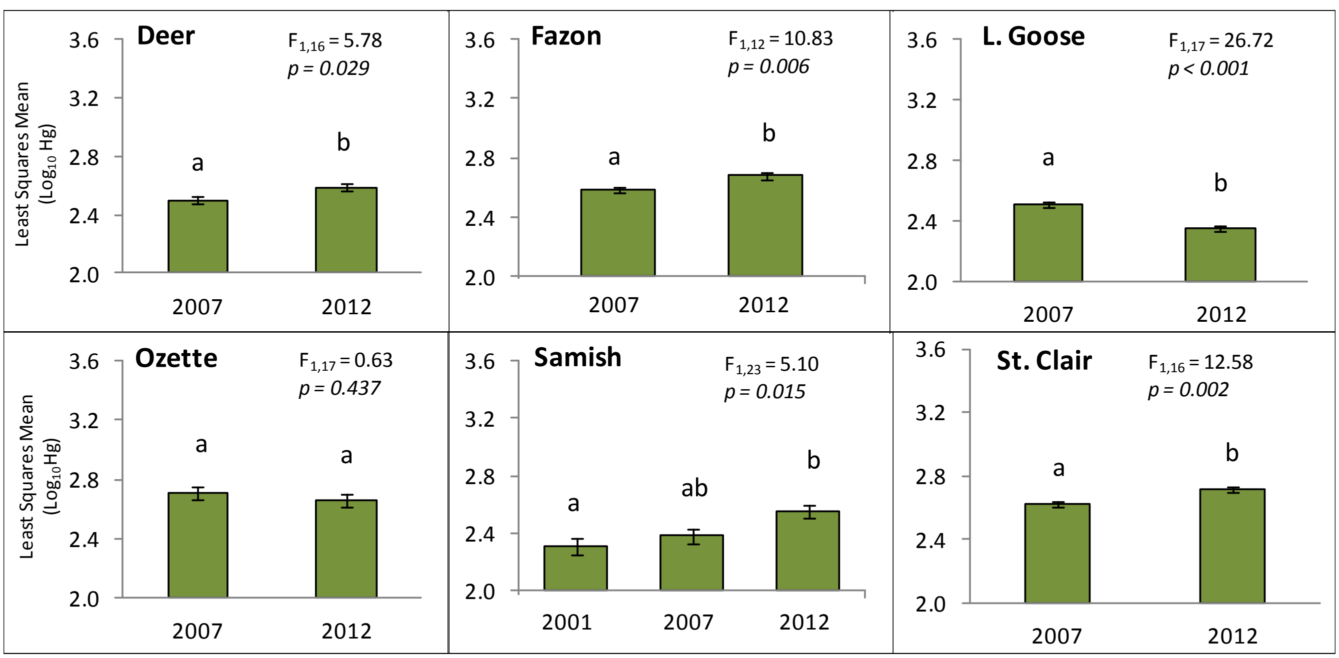


Figure 6. Results of ANCOVA and Bonferroni Post-Hoc Tests Comparing Mercury Levels in Bass Between Collection Years. *Error bars represent one standard error. F-ratios and p-values are ANCOVA results. A difference in letters indicates statistically significant difference in estimated group means (Bonferroni post hoc p value <0.05).*

Temporal Trends

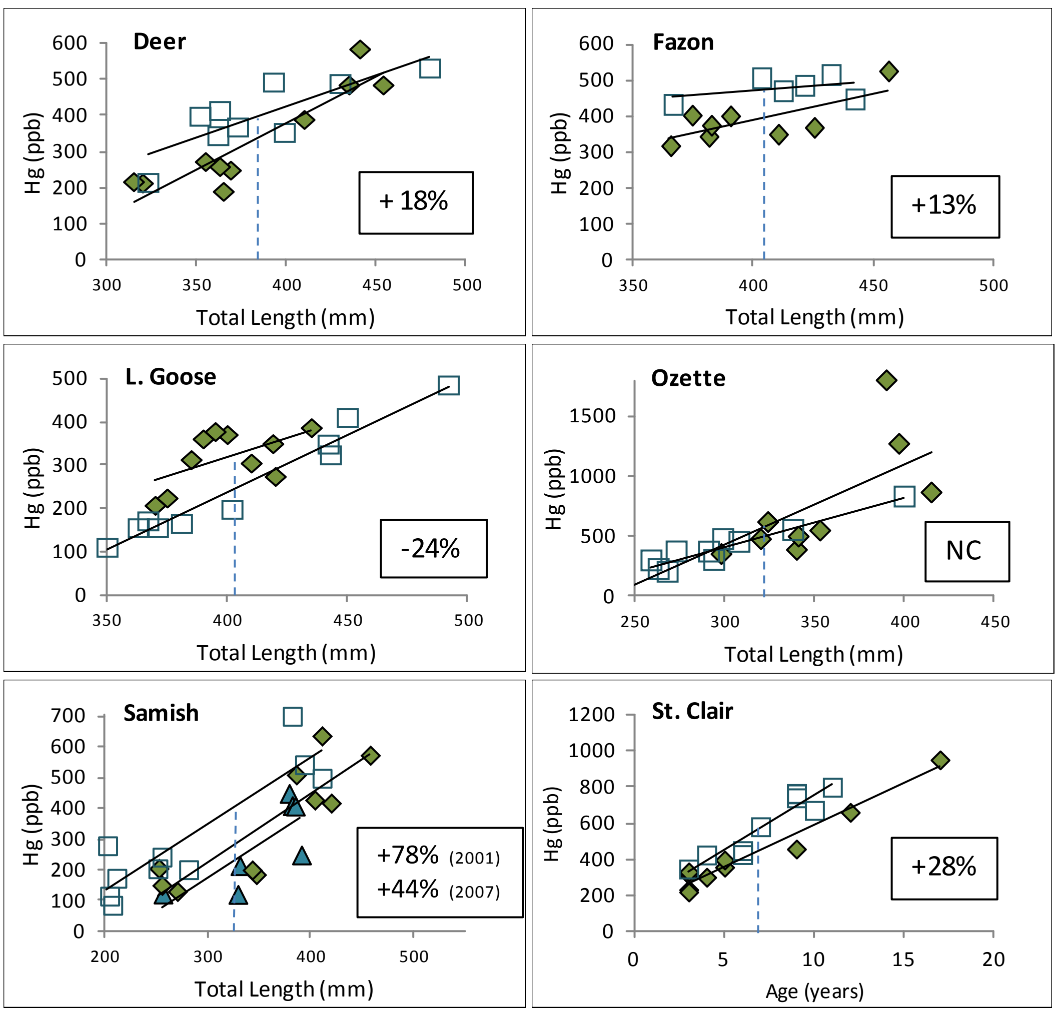
Mercury Levels in Standard-Size Bass

Standard-size bass mercury levels increased 18%, 13%, and 28% at Deer, Fazon, and St. Clair Lakes, respectively, between 2007 and 2012. Estimated standard-size concentrations in Lake Samish were 78% higher in 2012 than in 2001. In L. Goose Lake, 2012 concentrations were 24% lower than in 2007. The magnitude of change was calculated using percent change between mercury levels in standard-size bass (Figure 7). Standard size mercury values were estimated by least squares linear regression on raw data, using the average fish length (or age, for St. Clair) of each lake data set.

Mercury accumulation by top-predator fish species is determined by a number of different factors, including mercury loading, methylation rates, and food web dynamics. Sediment core records indicated rising mercury loading rates in Deer Lake and Lake St. Clair sediments from the 1970s through the mid-2000s (Furl, 2007b; Mathieu, 2013). Mercury in sediment core samples collected from Lake Ozette displayed no significant change during the same time period (Furl, 2007b). Lake Samish sediments showed a significant decrease in mercury levels from 1970 to 2010 (Mathieu and Friese, 2012). The contradictory trends between sediment core and fish tissue samples at Lake Samish suggest factors other than mercury loading may be responsible for the rising mercury levels in bass.

Water samples measured in 2007 and 2012 for alkalinity, DO, DOC, and pH, as well as antecedent precipitation values were similar for all lakes, revealing no change in factors measured that may influence methylation rates. Other parameters that have been shown to influence methylation, such as sulfate, were not measured in both years.

Relationships between fish lengths, weights, and ages were similar between collection years at each lake, indicating the differences in mercury are not likely attributable to changing growth rates. However, changes in fish communities and bass diets were not assessed, which can also influence mercury levels.



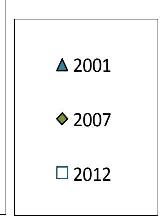


Figure 7. Mercury and Fish Length Data Sets for All Bass Collection Years. *Trendlines show linear regressions for each collection year. Percent change in linear regression standard-size bass mercury values at average fish length (or age) for each data set is included in boxes. Vertical dashed line: average fish length (or age) for data set. NC: not calculated.*

Temporal Trends

Composite Fish Samples

Mercury levels in composite samples collected in 2007 and 2012 are presented in Figure 8. Results from composite samples provide support for DOH's fish consumption advisories. They are not used to detect trends. Because of the smaller sample sizes, statistical trend tests are not applicable to composite data. Also, variability in species' sizes, diet, trophic position, and body composition prevent statistical comparisons across waterbodies and even within the same lake. The data presented in Figure 8 provide a qualitative view of mercury levels in species other than bass collected at the lakes in 2007 and 2012. Samples were paired based on average composite fish length (< 5% RPD).

Composites collected in 2012 had higher mercury concentrations than their respective 2007 sample in 8 out of 11 cases. 2012 mercury levels were higher in Deer Lake, Fazon Lake, and Lake St. Clair than in 2007. The direction of change was mixed at Lake Ozette. Northern pikeminnow composites from Ozette had lower mercury levels in 2012 than in 2007, whereas yellow perch from the lake had higher levels. Lake Samish northern pikeminnow samples contained very similar mercury levels between the two collection years.

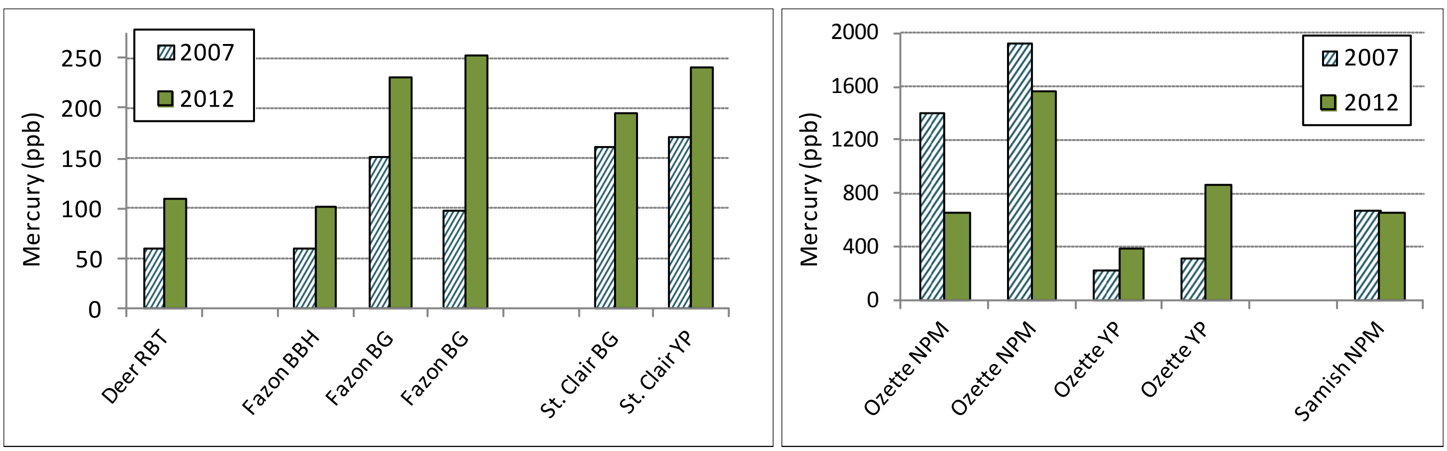


Figure 8. 2007 and 2012 Mercury Concentrations in Composite Samples Grouped by Similar Length.
RBT: rainbow trout; BBH: brown bullhead; BG: bluegill; NPM: northern pikeminnow; YP: yellow perch.

Conclusions

Ecology collected and analyzed individual bass and composite samples of other fish species from 6 lakes in 2012 as part of the eighth year of a long-term study. Fish were previously sampled from the waterbodies in 2007 and 2001 (Lake Samish only); this report summarizes changes in mercury levels over that time period.

Results of this study include the following:

- Statistical tests showed a significant increase in $\log_{10}$ mercury levels between 2007 and 2012 at 3 of the 6 lakes (Deer, Fazon, and St. Clair). In 2012, mercury levels of standard-size bass were 18%, 13%, and 28% higher than those measured in 2007.
- Log_{10} mercury concentrations increased significantly between 2001 and 2012 at Lake Samish. Standard-size bass mercury levels increased by 78% between 2001 and 2012 in the lake.
- Log_{10} mercury levels in bass from L. Goose Lake were significantly lower in 2012 than in 2007. Standard-size bass mercury levels decreased 24% from 2007.
- No significant change was found in Lake Ozette bass mercury levels between 2007 and 2012.
- Composite fish samples collected in 2012 were higher in mercury concentrations than similar-sized 2007 samples in 8 out of 11 cases.
- Two individual largemouth bass samples exceeded the NTR regulatory criterion of 770 ppb, collected from Lakes Ozette and St. Clair. Three composite samples (northern pikeminnow and yellow perch) collected from Lake Ozette were also above the NTR regulatory criterion.
- Over half (55%) of individual bass and 36% of composite fish mercury concentrations were above the EPA Recommended Criterion for human health (300 ppb).

Recommendations

The authors recommend the following:

- The Washington State Department of Health should review the results of this report and consider the information when making or updating fish consumption advisories.
- Ecology should include this data in the next water quality assessment. Five samples violated the state regulatory criterion for mercury.
- A follow-up study may be considered to evaluate the rising mercury trend in fish tissue in Lake Samish. Increases in this lake were much higher than at others, and samples often exceeded the EPA recommended criterion. Further study may include investigation into mercury loading rates (e.g., high-resolution sediment cores, suspended sediment collections near major tributaries and outfalls, and sampling mercury and TSS in surface water tributaries and outfalls) and/or assessing bass diet and community structure.
- Methyl mercury may be added to the list of water sample analytes at a subset of sampling locations in 2014 (Lakes Whatcom, Failor, and Pierre). This data will strengthen our knowledge of factors influencing mercury levels in fish in Washington State.

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Department of Ecology Contacts

Lead Author: Callie Mathieu
callie.mathieu@ecy.wa.gov
Environmental Assessment Program
P.O. Box 47600
Olympia, WA 98504-7600

Communications Consultant
Phone: 360-407-6764

Washington State Department of Ecology - www.ecy.wa.gov/
Headquarters, Olympia: 360-407-6000

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